Supplementary information

Inhibition of transition metals dissolution in cobalt-free cathode with ultrathin robust interphase in concentrated electrolyte

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Supplementary Table 1. Structure parameters of $Li_{1.2}Ni_{0.15}Fe_{0.1}Mn_{0.55}O_2$, space group: $R\bar{3}m$

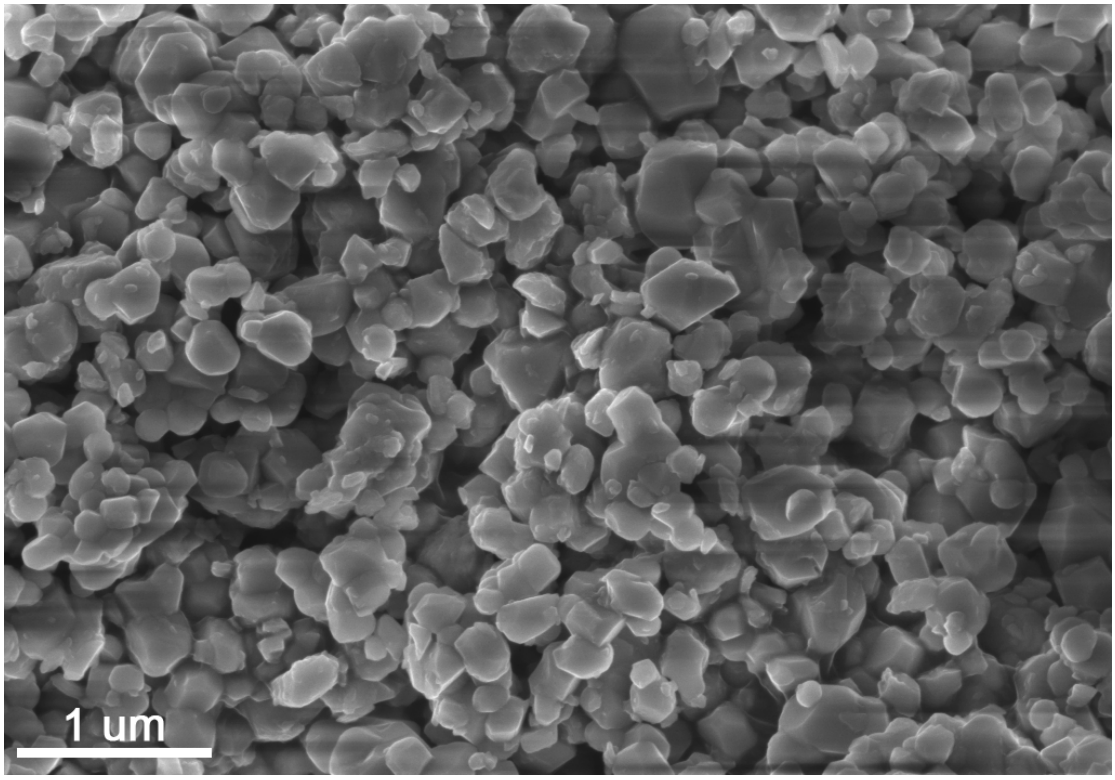
	Lattice parameters						
Sample	a（Å）	c（Å）	c/a	v（$Å^3$）	I_{003}/I_{104}	Rp(%)	Rwp（%）
$Li_{1.2}Ni_{0.15}Fe_{0.1}Mn_{0.55}O_2$	2.8676	14.2760	4.9784	101.6654	1.45	7.61	8.73

Supplementary Table 2. Chemical compositions acquired from ICP-OES analysis

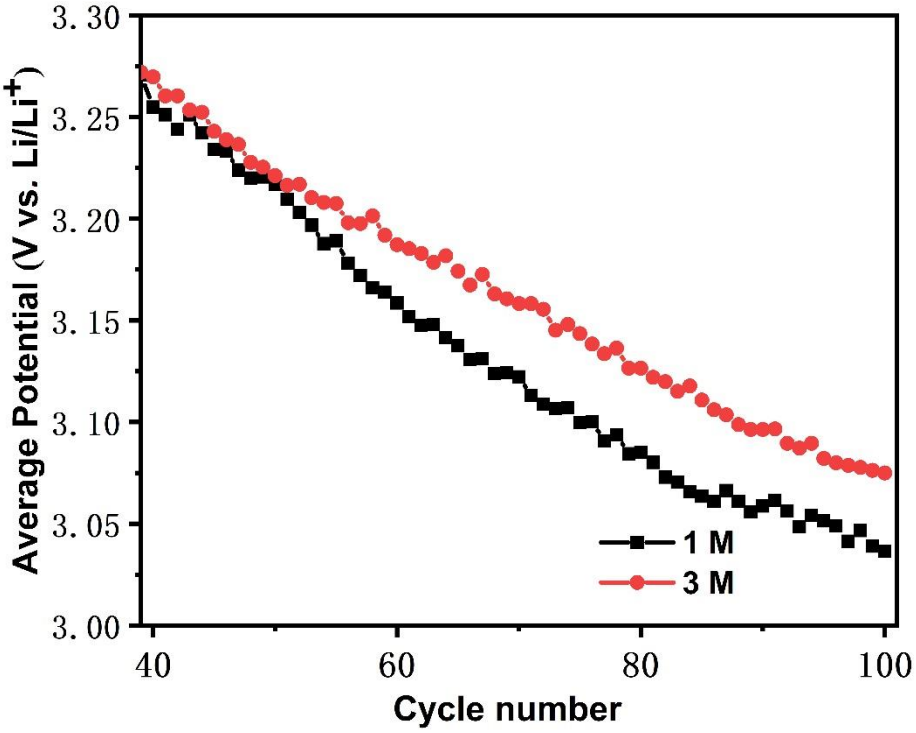
Sample	Theoretical molar content of Li:Ni:Fe:Mn	Experimental molar content of Li:Ni:Fe:Mn
$Li_{1.2}Ni_{0.15}Fe_{0.1}Mn_{0.55}O_2$	1.20:0.15:0.10:0.55	1.199:0.159:0.100:0.542

Supplementary Table 3. Physicochemical Properties of electrolytes at 25℃

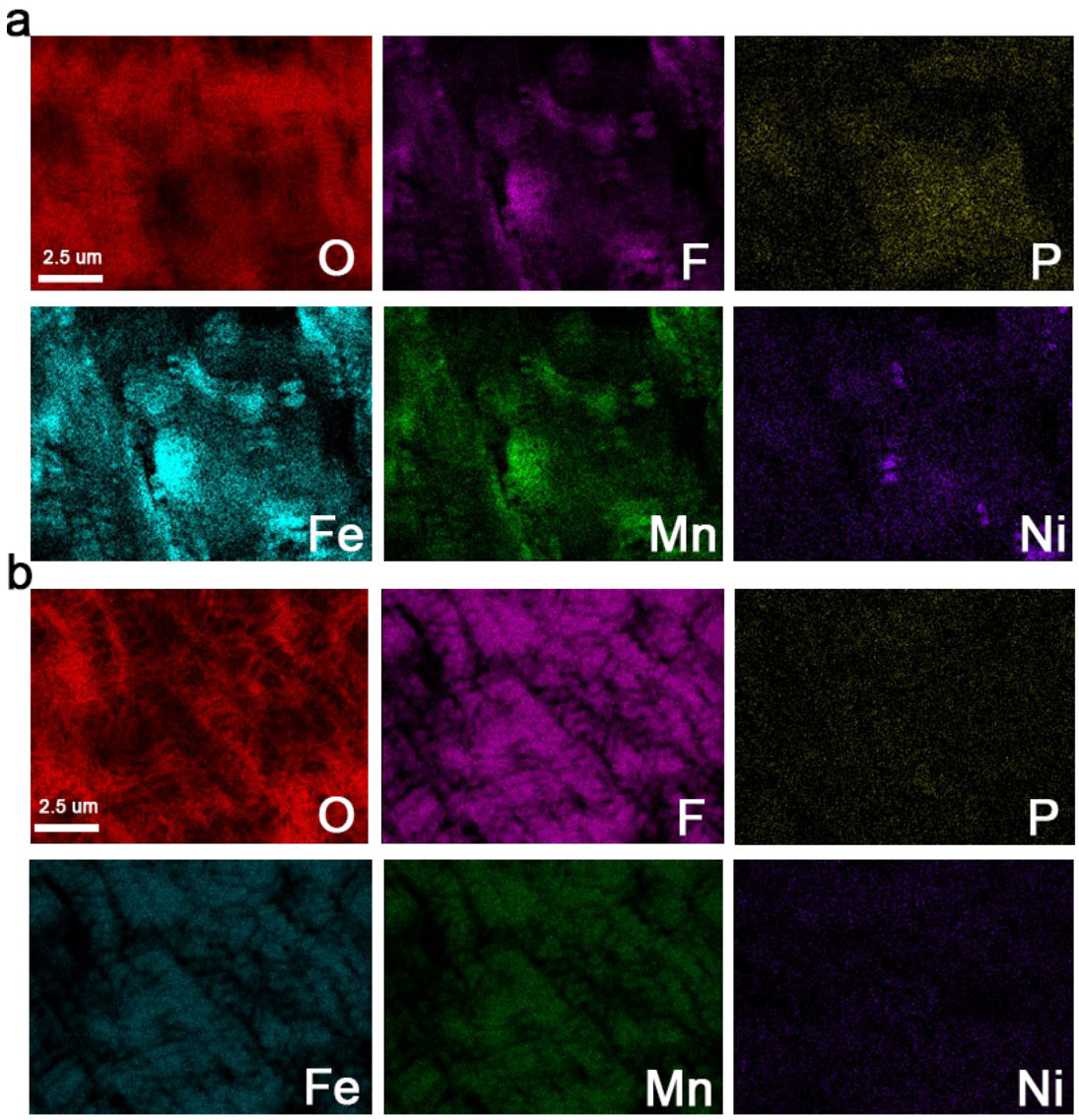
Concentration ($mol \cdot dm^{-3}$)	Density ($g \cdot cm^{-3}$)	Viscosity (mPa·s)	Conductivity ($mS \cdot cm^{-1}$)
1	1.188	3.187	10.48
3	1.327	19.809	5.30
4	1.432	54.434	2.71
5	1.509	99.471	1.57



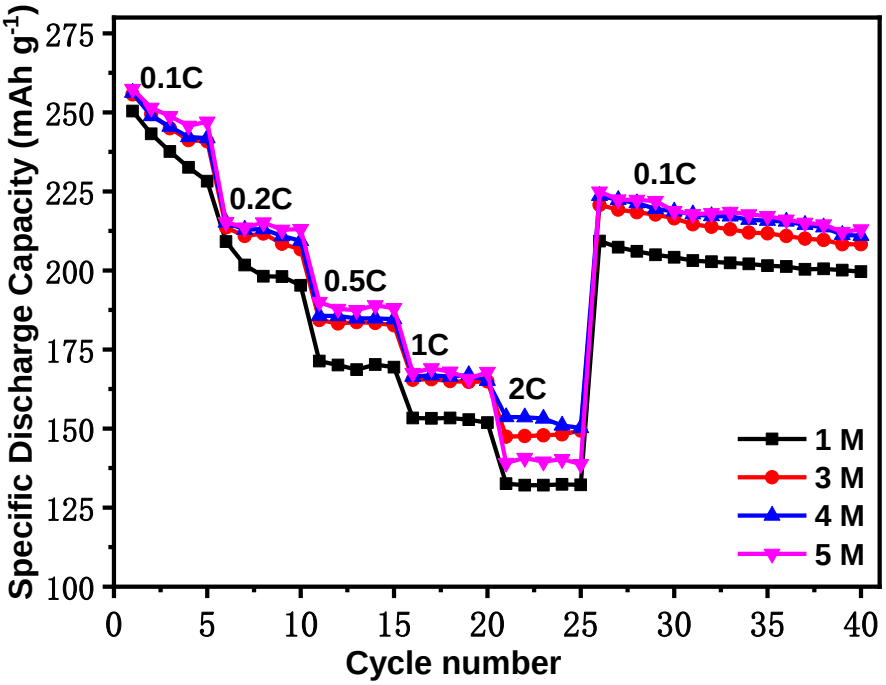
Supplementary Fig. 1. SEM image of pristine sample.



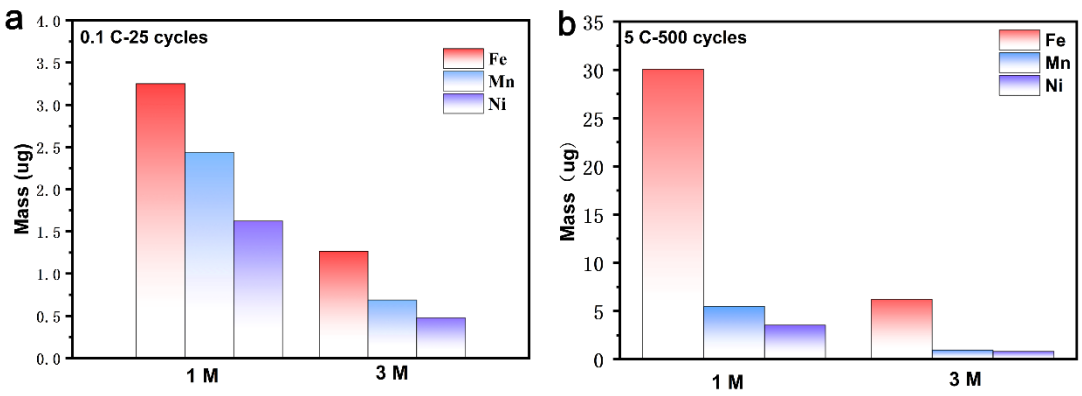
Supplementary Fig. 2. The corresponding zoom of Fig. 3b.



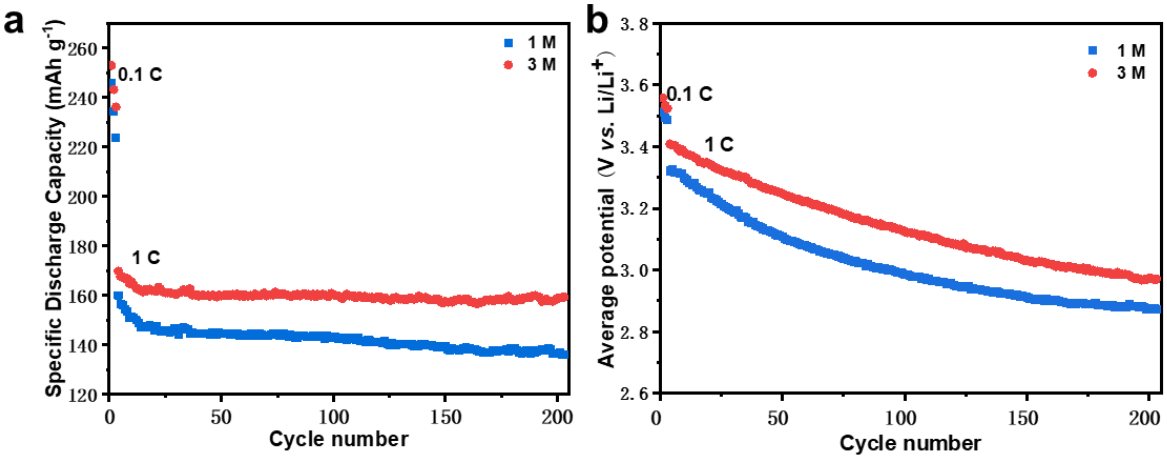
Supplementary Fig.3. The SEM-EDS spectra of 100 cycled lithium counter electrode. The SEM-EDS spectra of Lithium counter electrode cycled 100 cycles in 1 M electrolyte (a), and 3 M electrolyte (b), respectively.



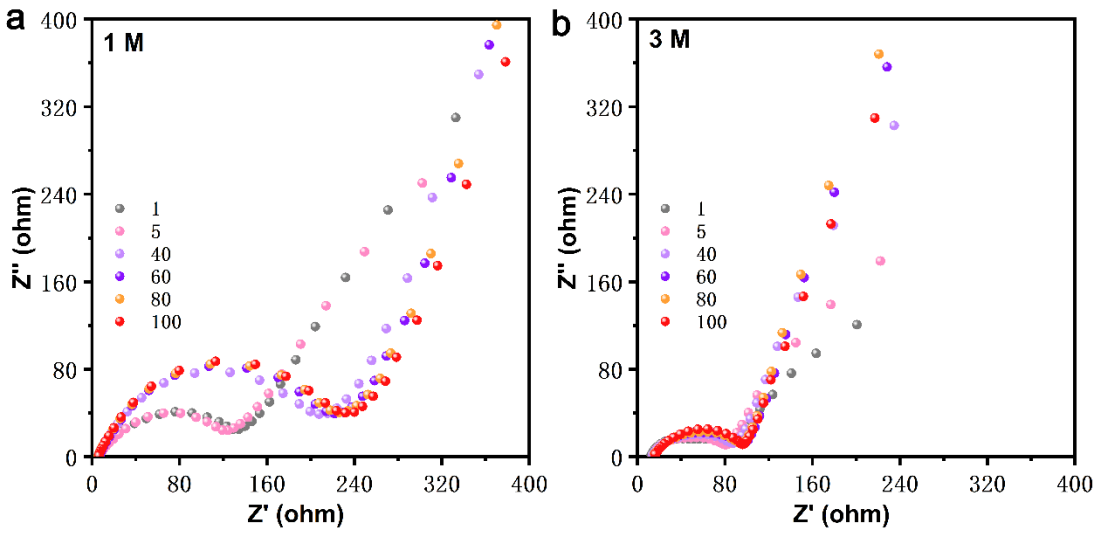
Supplementary Fig.4. Specific discharge capacity in various electrolytes at various C-rates



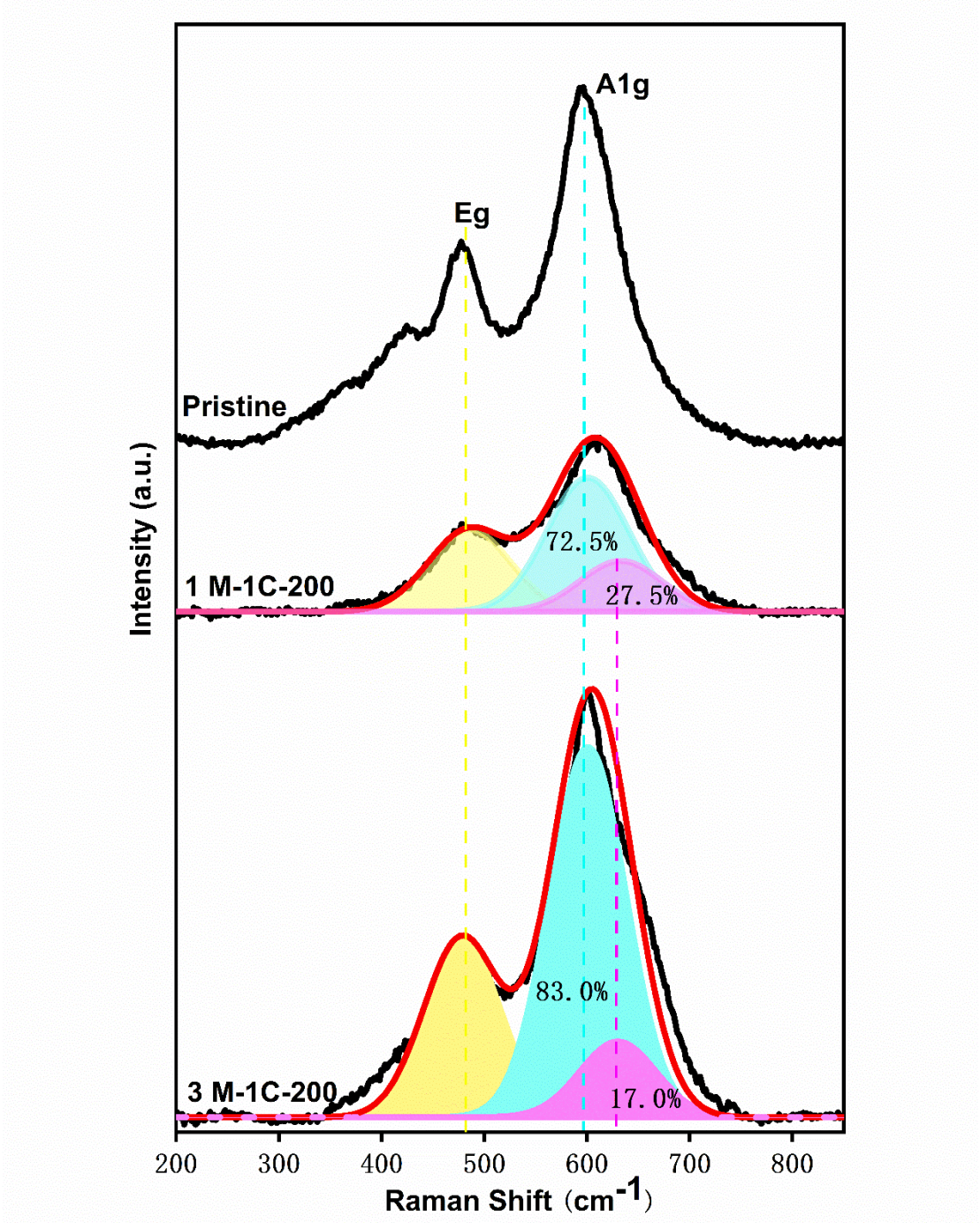
Supplementary Fig. 5. The amount of deposited transition metals on lithium metal. The amount of transition metals deposited on lithium metal counter electrode after 25 cycles at 0.1 C (a) and 500 cycles at 5 C (b), respectively.



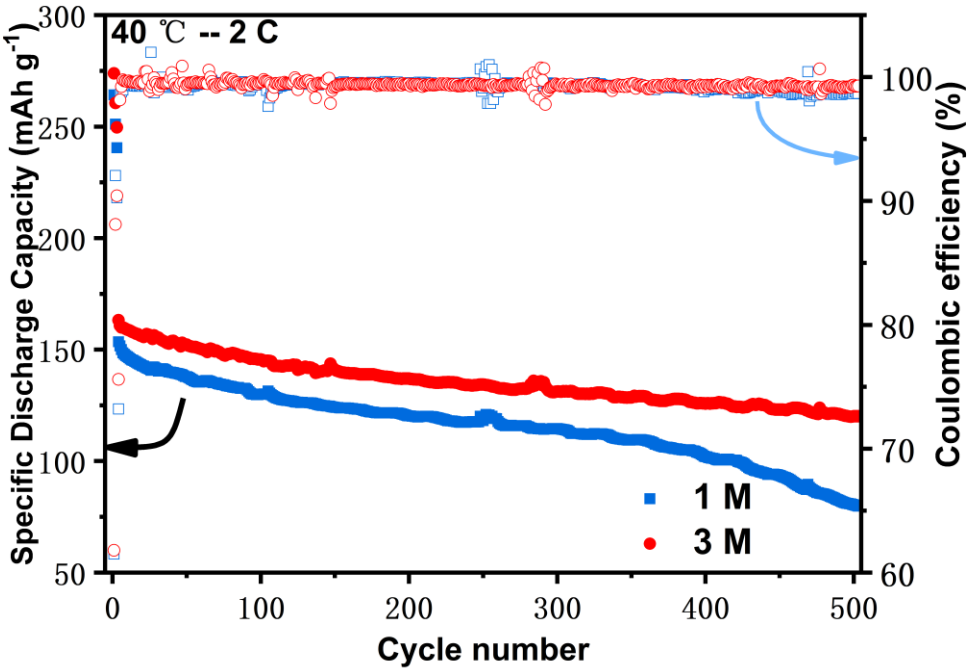
Supplementary Fig. 6. The electrochemical performance of LNFMO at 1 C. (a) The cycle performance at 1C in 1M and 3M electrolyte at 25 °C. (b) The corresponding average discharge potential at 1C in 1M and 3M electrolyte at 25 °C.



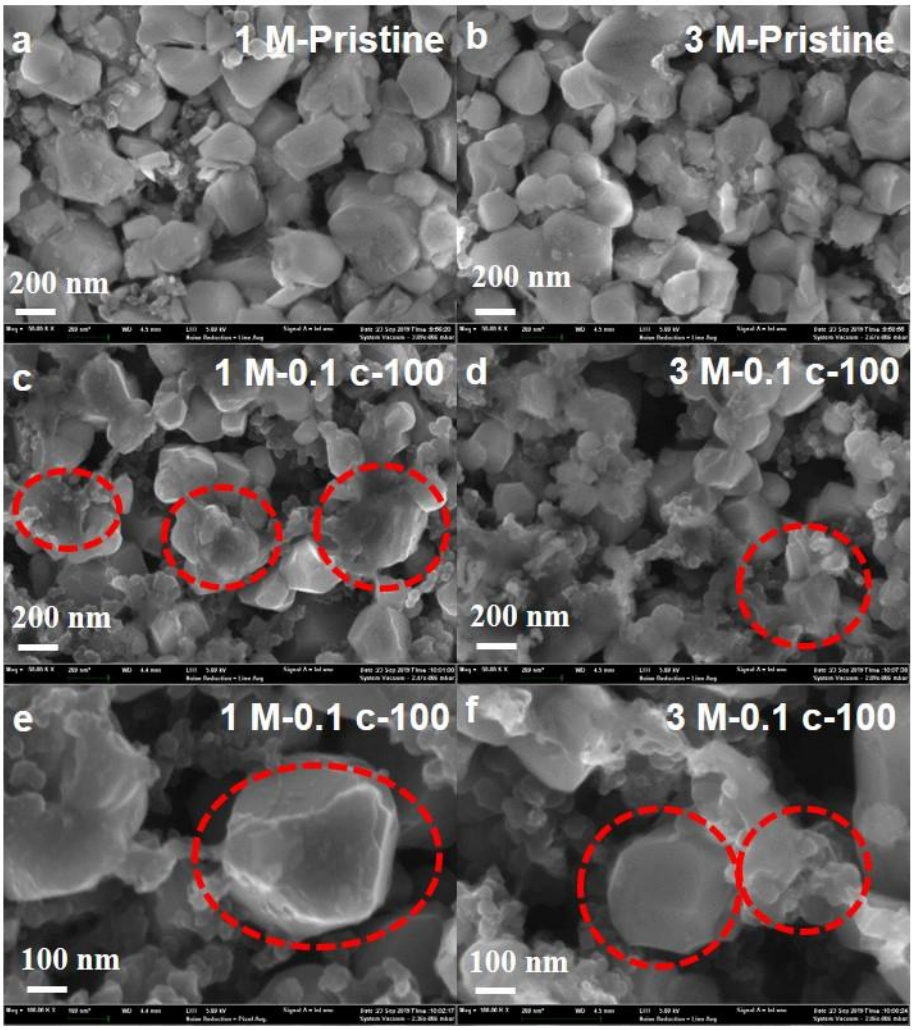
Supplementary Fig. 7. The dependence of EIS spectra of electrode with cycle number. The dependence of EIS spectra of LNFMO electrode discharged to the 50% state of charge at 1 M electrolyte (a) and 3 M electrolyte (b) with cycle number.



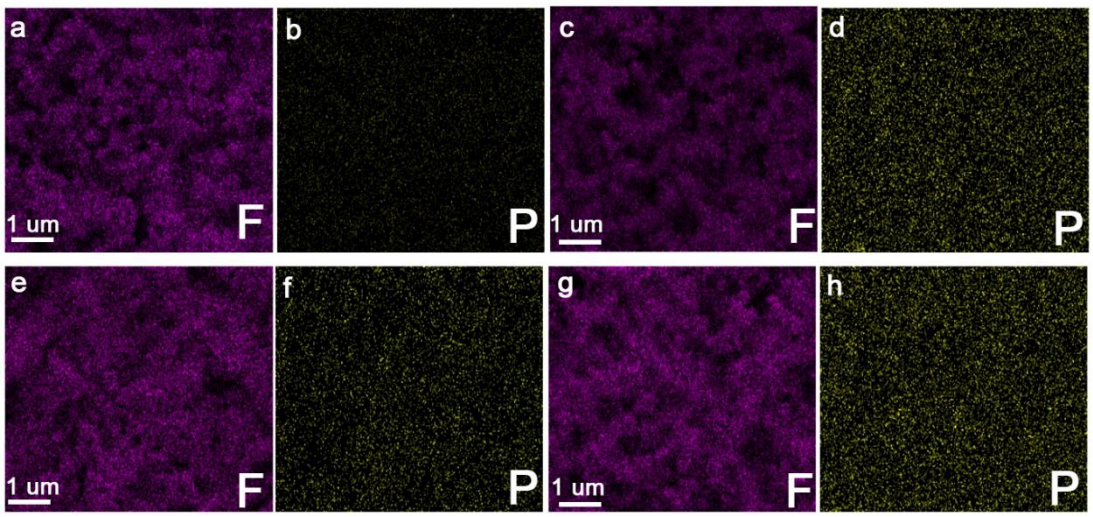
Supplementary Fig. 8. The Raman spectra of pristine LLNFMO and the electrodes after 200 cycles at 1C in 1 M and 3 M electrolyte.



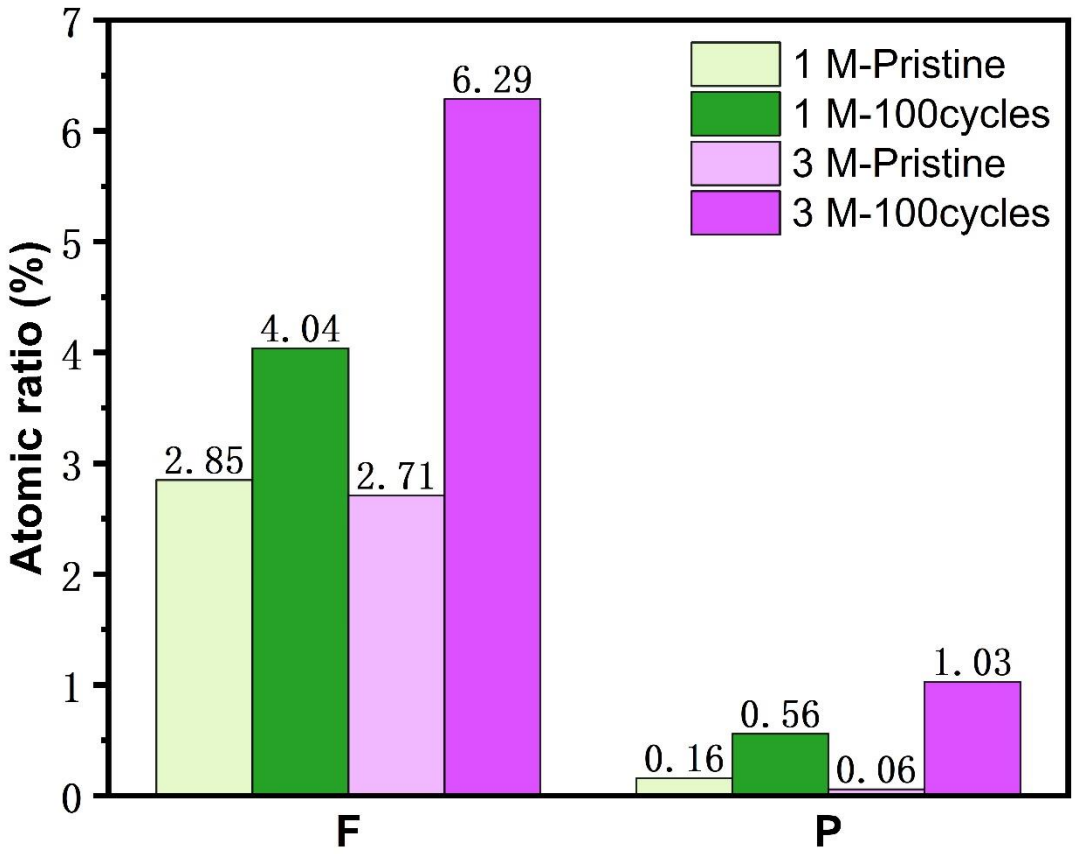
Supplementary Fig.9. The 2 C rate cycle performance in 1 M and 3 M electrolytes at elevated temperature (40 °C).



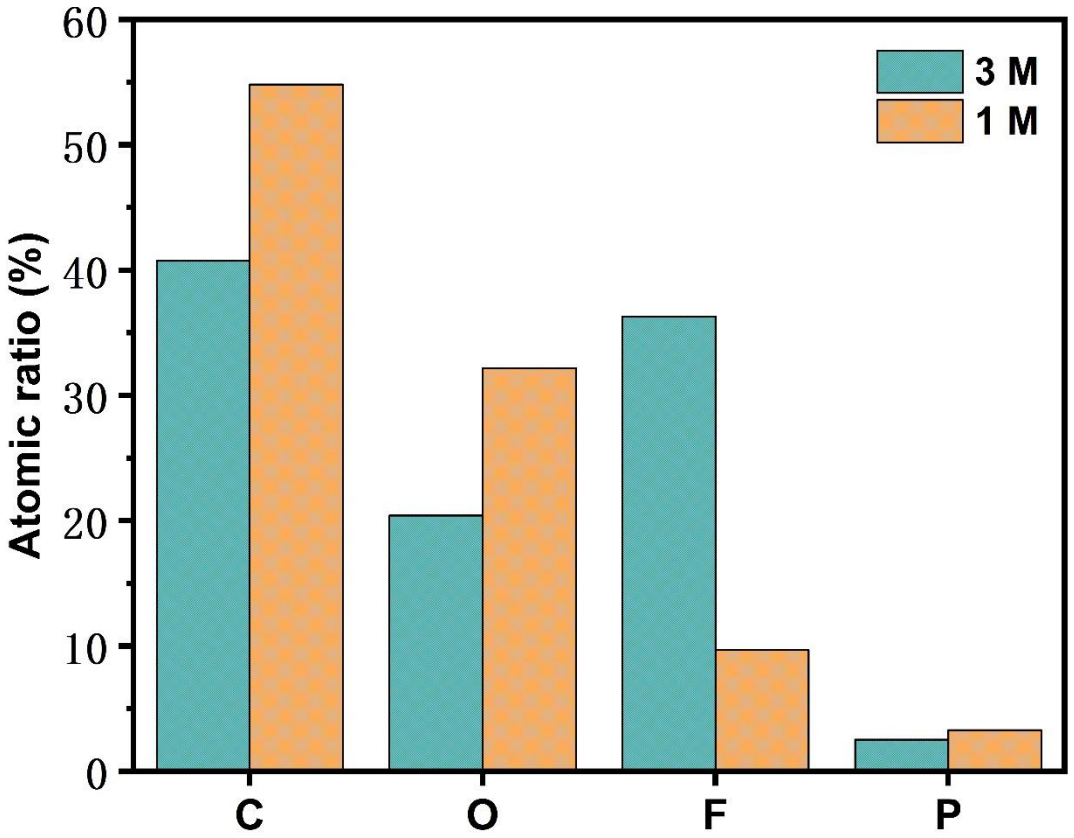
Supplementary Fig.10. The SEM images of electrode. The SEM images of the pristine electrode The SEM images of the at 0.1C in 1 M (a) and 3 M electrolytes (b); the SEM images of the 100 cycled electrode at 0.1 C in 1 M (c,e) and 3 M (d,f) electrolyte.



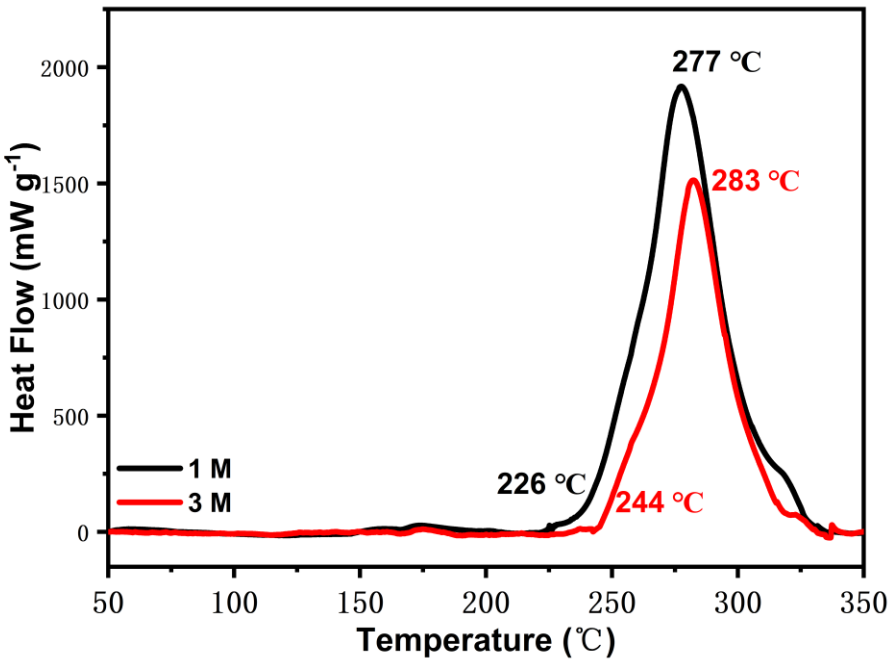
Supplementary Fig.11. EDS mapping of F、P for the pristine and 100 cycled at 0.1C .(a, b) The pristine of LLNFMO Shelved for 24 hours in 1 Melectrolyte; (c, d) LLNFMO after 100 cycles in 1 M electrolyte at 0.1C; (e, f) the pristine of LLNFMO shelved for 24 hours in 3 M electrolyte; (g, h) LLNFMO after 100 cycles in 3 M electrolyte at 0.1C .



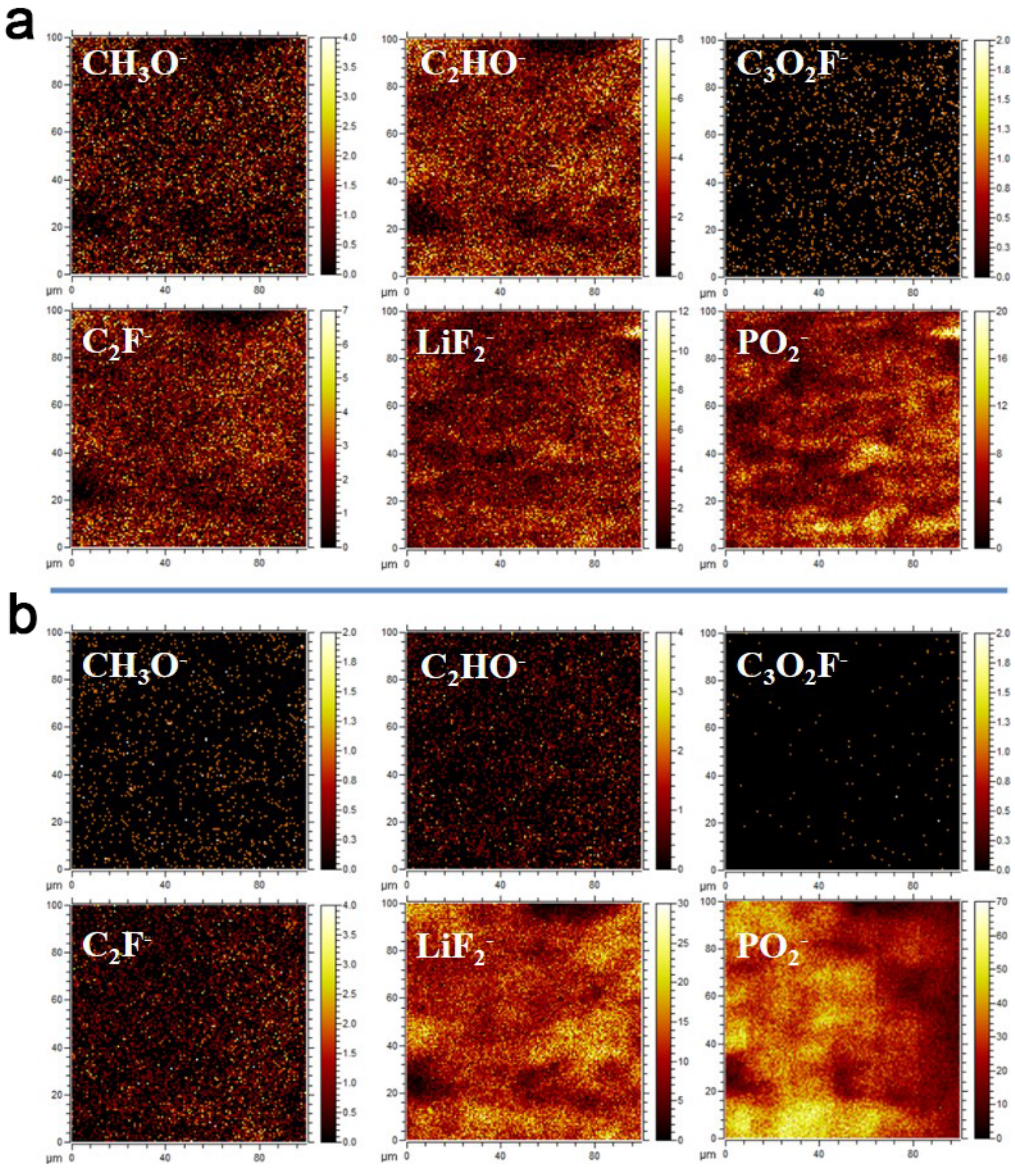
Supplementary Fig. 12. The atomic ratio of F and P on electrode surface. The atomic ratio of F and P on the surface of pristine (shelved for 24 hours in coin cell) and 100 cycled at 0.1C in 1 M and 3 M electrolytes.



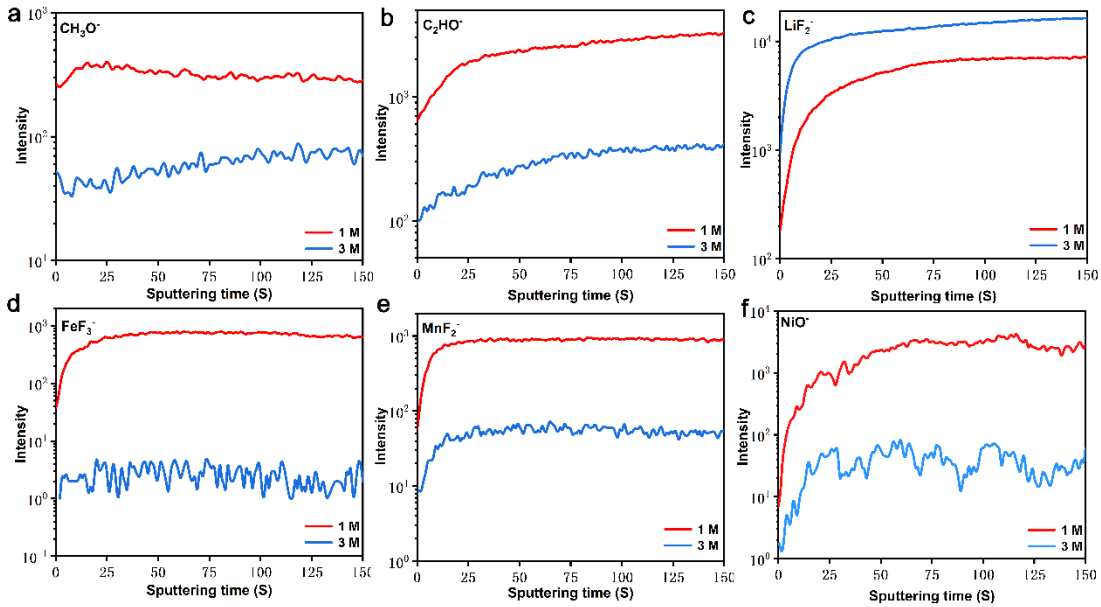
Supplementary Fig. 13. The element atomic ratio of cycled electrode. The atomic ratio of C, O, F, and P on the surface of electrode cycled in 1 M and 3 M electrolyte at 0.1C for 100 cycles obtained by XPS spectra.



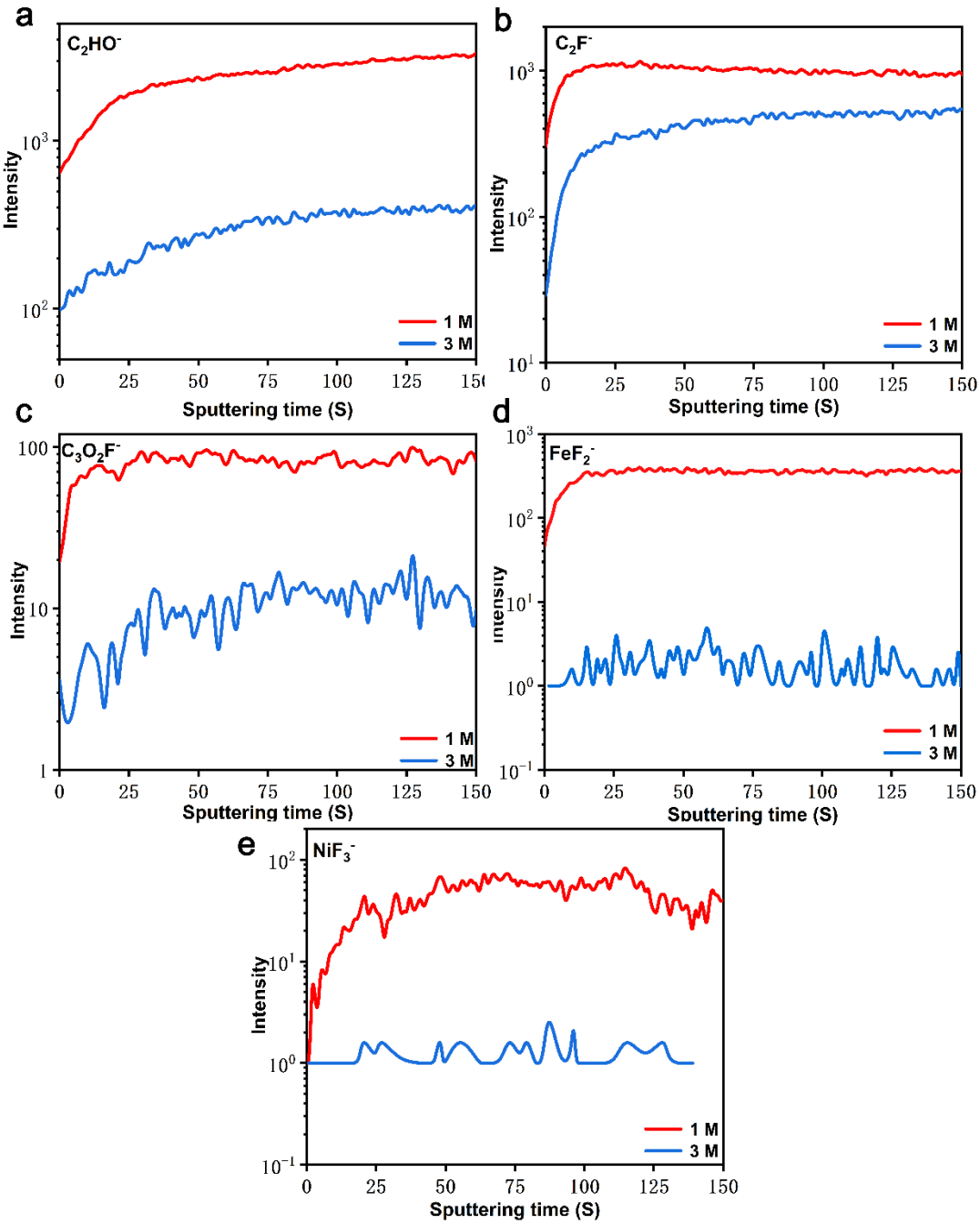
Supplementary Fig. 14. **DSC profile of electrode.** The DSC profile of LNFMO electrode after the first charging to 4.8V in 1 M and 3 M electrolyte.



Supplementary Fig. 15. TOF-SIMS chemical maps of several typical second ion fragments. TOF-SIMS chemical maps of several typical second ion fragments with 25s of sputtering on electrodes cycled in 1 M electrolyte (a) and 3 M electrolyte (b), respectively.



Supplementary Fig. 16. Corresponding depth profiling of second ion fragments on electrodes. The corresponding depth profiling of CH_3O^- (a) , C_2HO^- (b), LiF_2^- (c), FeF_3^- (d), MnF_2^- (e), and NiO^- (f) second ion fragments with 150s of sputtering on electrodes cycled in 1 M and 3 M electrolyte, respectively.



Supplementary Fig. 17. Depth profiling of second ion fragments of interest on electrodes. The depth profiling of C_2HO^- (a) , C_2F^- (b), $C_3O_2F^-$ (c), FeF_2^- (d), and NiF_3^- (e) second ion fragments with 150s of sputtering on electrodes cycled in 1 M and 3 M electrolyte, respectively.